Resilient sealing materials for solid oxide fuel cells

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NT42175 (DE-FG26-04NT42175)

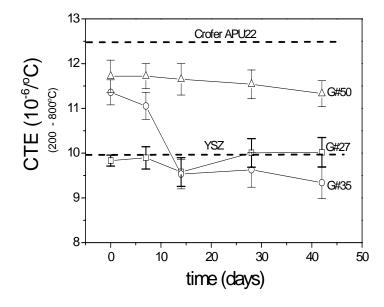
Period of performance: Oct. 1, 2004-Sept. 30, 2006

Objective

The objective of this Phase I program is to develop, characterize, fabricate, and test a hermetic solid oxide fuel cell (SOFC) seal based on a thermochemically-stable glass-ceramic system. The seal will be fabricated below 900°C and will be tested for under cell operating environments, including temperatures in the range 750-800°C.

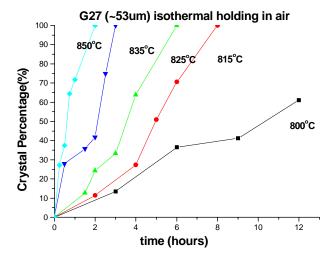
Accomplishments to Date

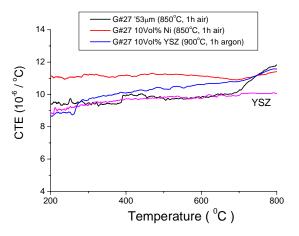
We have developed and characterized glass compositions that, upon sealing and crystallization, possess desirable thermo-mechanical properties for SOFC sealing materials. The glasses are based on the alkaline earth silicate system, with other oxides added to control crystallization behavior and to tailor thermal properties. Certain compositions possess stable thermal expansion characteristics that are compatible with other materials used in SOFC stacks. For example, the figure to the right shows the coefficients of thermal expansion (CTE) for three crystallized glasses (designated G#27, G#35, and G#50) held at 800°C for up to 42 days. The CTEs of G#27 and G#50 are relatively constant, whereas the CTE for G#35 decreases over the first ten days. The latter behavior is due to a change in the distribution of crystalline phases that constitute the glass-ceramic. The stability of the CTE's for G#27 and G#50 makes them good candidates for further sealing experiments.



We have used differential thermal analyses (DTA) to study the crystallization kinetics of the sealing glasses. The exothermic crystallization peak area in a DTA scan is related to the fraction of residual glass in a sample that has undergone an isothermal heat treatment. By characterizing these peaks as a function of heat-treatment time and information temperature, (including crystallization kinetics activation energies) can be obtained. The figure to the right summarizes a series of DTA experiments performed on the SOFC sealing glass #27. Other experiments shown) (not reveal information about the effects of particle size and the presence of a second phase

(composite seals) on crystallization kinetics.





Finally, we are using the sealing glasses developed in this project as the bases for

producing composite seals. Glass powders have been mixed with a variety of second phases, including metals and ceramics, and thermal properties and phase stabilities have been evaluated. The figure to the left compares the CTE characteristics of G#27 undoped, with 10 vol% Ni powder, and 10vol% YSZ powder, to the CTE for YSZ (Ystabilized zirconia). Tape-cast sealing performs have been prepared and used for fabricating test seals with SOFC materials.

The tasks presently underway are summarized below:

- 1. Develop test fixtures to evaluate the effects of thermal cycling on the hermeticity of SOFC seals fabricated from glass and glass-ceramic composite seals.
- 2. Complete the long-term studies of phase and property stabilities of sealing materials.
- 3. Complete the DTA studies of crystallization kinetics.

Publications (since October 1, 2004):

- S.T. Reis, R.K. Brow, "Designing Sealing Glasses for Solid Oxide Fuel Cells," Proceedings of the ASM Materials Solution Conference, Fuel Cells: Materials, Processing and Manufacturing Technologies, Columbus, OH Oct. 18-20, 2004.
- S.T. Reis, R.K. Brow. P. Jasinski, and T. Zhang, "Properties of Glass-Ceramic for Solid Oxide Fuel Cells," proceedings of the 3rd International Symposium on Solid Oxide Fuel Cells, 30th International Conference & Exposition on Advanced Ceramics & Composites, Cocoa Beach, FL, Jan. 22-27, 2006.; accepted for publication by the American Ceramic Society, 3/14/06

Patent Actions (since October 1, 2004):

• R.K. Brow, S. T. Reis, G. M. Benson, "Glass and glass-ceramics for solid oxide fuel cell hermetic seals," US Patent Application, UM Disclosure No. 04UMR023 entitled "Glass and Glass-Ceramic Sealant Compositions," filed January 2005.

Presentations (since October 1, 2004):

- R. K. Brow and S.T. Reis, "Designing Sealing Glasses for Solid Oxide Fuel Cells," ASM Materials Solution Conference, Fuel Cells: Materials, Processing and Manufacturing Technologies, Columbus, OH Oct. 18-20, 2004 (INVITED).
- T. Zhang, S. T. Reis, and R. K. Brow, "Glass-ceramic seals for solid oxide fuel cells," NSF I-U Center for Glass Research, Semi-annual meeting, Sarasota, FL, January 19-21, 2005.
- S. T. Reis*, R. K. Brow, and P. Jasinski, "Developing Glass Seals for Solid Oxide Fuel Cells," 2nd International Symposium on Solid Oxide Fuel Cells: Materials and Technology, 29th International Cocoa Beach Conference and Exposition on Advanced Ceramics and Composites, Cocoa Beach, FL, January 23-28, 2005.
- R.K. Brow*, "Glass Seals for Solid Oxide Fuel Cells," Iowa State Materials Science & Engineering Seminar, Ames, IA, March 3, 2005. (INVITED)
- Teng Zhang*, S. T. Reis, and R. K. Brow, "Glass Seals for Solid Oxide Fuel Cells," 107th Annual Meeting of the American Ceramic Society, Baltimore, MD, April 10-13, 2005.
- R.K. Brow, "Thermochemically stable sealing materials for solid oxide fuel cells," Solid State Energy Conversion Alliance 6th Annual Workshop, Pacific Grove, CA, Apr. 18-21, 2005. (INVITED)
- R.K. Brow, "Sealing Glasses for Solid Oxide Fuel Cells," 17th University Conference on Glass Science and 1st International Materials Institute Workshop on "New Functionality in Glasses", Penn Stater Conference Center Hotel, State College, PA, June 26-30, 2005. (INVITED)
- T. Zhang*, S. T. Reis, R. K. Brow, and C.S. Ray, "Crystallization Studies of SOFC Sealing Glasses," 3rd International Symposium on Solid Oxide Fuel Cell: Materials and Technology, 30th International Conference & Exposition on Advanced Ceramics & Composites, Cocoa Beach, FL, Jan. 22-27, 2006.
- S.T. Reis*, R.K. Brow, and T. Zhang, "Glass-Ceramic Seals for Solid Oxide Fuel Cells: Thermo-Phase Stability," 3rd International Symposium on Solid Oxide Fuel Cell: Materials and Technology, 30th International Conference & Exposition on Advanced Ceramics & Composites, Cocoa Beach, FL, Jan. 22-27, 2006.